

# Spatial and indoor/outdoor gradients in urban concentrations of ultrafine particles and PM<sub>2.5</sub> mass and chemical components

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3 **Spatial and Indoor/Outdoor Gradients in Urban**  
4 **Concentrations of Ultrafine Particles and PM<sub>2.5</sub> Mass**  
5 **and Chemical Components**  
6  
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8

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30 **ABSTRACT**

31 In order to investigate relationships between outdoor air pollution and concentrations indoors, a  
32 novel design of experiment has been conducted at two sites, one heavily trafficked and the other  
33 residential. The novel design aspect involves the introduction of air directly to the centre of an  
34 unoccupied room by use of a fan and duct giving a controlled air exchange rate and allowing an  
35 evaluation of particle losses purely due to uptake on indoor surfaces without the losses during  
36 penetration of the building envelope which affect most measurement programmes. The rooms were  
37 unoccupied and free of indoor sources, and consequently reductions in particle concentration were  
38 due to deposition processes within the room alone. Measurements were made of indoor and  
39 outdoor concentrations of PM<sub>2.5</sub>, major chemical components and particle number size distributions.  
40 Despite the absence of penetration losses, indoor to outdoor ratios were very similar to those in  
41 other studies showing that deposition to indoor surfaces is likely to be the major loss process for  
42 indoor air. The results demonstrated a dramatic loss of nitrate in the indoor atmosphere as well as a  
43 selective loss of particles in the size range below 50 nm, in comparison to coarser particles.  
44 Depletion of indoor particles was greater during a period of cold weather with higher outdoor  
45 concentrations probably due to an enhancement of semi-volatile materials in the outdoor particulate  
46 matter. Indoor/outdoor ratios for PM<sub>2.5</sub> were generally higher at the trafficked site than the  
47 residential site, but for particle number were generally lower, reflecting the different chemical  
48 composition and size distributions of particles at the two sites.

49

50 **Keywords:** Indoor-outdoor air; deposition; PM<sub>2.5</sub>; nanoparticles

51

52

## 53 1. INTRODUCTION

54 Atmospheric aerosol has been documented to cause increased mortality, morbidity, decreased lung  
55 function and other adverse effects upon health (Beelen et al., 2014; Raaschou-Nielsen et al., 2013),  
56 although there is considerable uncertainty about which physical and/or chemical characteristics of  
57 particulate matter (PM) are most important as determinants of health effects (Brunekreef and  
58 Holgate, 2002, REVIHAAP, 2013). Recently, toxicological and epidemiological studies have  
59 focused on health effects from exposure to ultrafine particles (UFP, particles with diameter <100  
60 nm) due to their toxicity and ability to penetrate deeply in the human lung (Peters et al., 2011; Hoek  
61 et al., 2010; von Klot et al., 2005)

62  
63 Traffic is the main source of fine and ultrafine particles and a principal determinant of the spatial  
64 pattern of air pollution within urban areas. While exposure to PM from vehicular emissions has  
65 been demonstrated to have detrimental impacts on human health (HEI, 2010) epidemiological  
66 evidence of adverse health effects associated with residential proximity to traffic is still limited.  
67 Some studies have shown a higher prevalence of respiratory symptoms (e.g. Delfino et al., 2014),  
68 especially in children (Gasana et al, 2012) but others did not find any effects (Badaloni et al., 2013;  
69 Macintyre et al., 2014).

70  
71 A key issue in studies on residential proximity to traffic is exposure assessment. Substantial efforts  
72 have been made in this field and a significant improvement has been reached with Land Use  
73 Regression Models (LUR) which make use of a spatially dense network of measured air pollution  
74 concentrations together with predictor variables such as population density, land use, and various  
75 traffic related variables to estimate outdoor air pollution concentrations within urban areas (Hoek et  
76 al., 2008a). However, exposure to pollutants takes place mainly indoors (Monn, 2001) and  
77 assessment of both indoor and outdoor variability of concentrations and characteristics of particles

78 are of primary importance to better understand the way residential proximity to traffic sources could  
79 affect human health.

80  
81 The relationships between indoor levels due to outdoor and indoor sources vary between cities,  
82 regions and countries due to differences in factors that can influence the indoor levels, e.g. climate,  
83 building characteristics, human activity, ventilation and heating systems (Monn, 2001; Nazaroff,  
84 2004; Ashmore and Dimitroulopoulou, 2009). However, it is reasonable that indoor sources could  
85 be considered a relevant, sometimes dominant, white noise superimposed upon spatial variation of  
86 exposure due to outdoor air that infiltrates indoors. Thus, it is particularly important to assess the  
87 penetration characteristics of particles into indoor environments, and the differences in physical and  
88 chemical properties of particles of outdoor origin.

89  
90 There are two possibilities to assess the mean differences in exposure due to proximity to traffic.  
91 The first is to measure a large number of indoor environments filtering out the effects of indoor  
92 sources and personal habits (Fuller et al., 2013; Spinazzè et al., 2013) . This type of study has the  
93 drawback of a strong limitation of the number of indoor environments which can be studied, and of  
94 the measurement duration of air pollutants, personal habits and air exchange rates. The other is to  
95 compare uninhabited indoor environments with characteristics and air exchange rates typical of  
96 residential settings. Very few studies have been conducted using this second approach (Schneider et  
97 al., 2004).

98  
99 In this paper we present the results of an experimental study carried out in a highly polluted city in  
100 Northern Italy following this second approach. Indoor and outdoor PM<sub>2.5</sub> mass and chemical  
101 composition as well as the size distribution of ultrafine particle have been contemporaneously  
102 measured at two sites with very different characteristics in relation to proximity to traffic sources.  
103 The objectives of the study were to compare indoor/outdoor (I/O) ratios of particulate pollutants in  
104 two similar unoccupied buildings with very different proximity to traffic, and to quantify I/O ratios

105 when air exchange ratios were well defined and penetration losses were eliminated by experiment  
106 design.

## 107 108 **2. METHODS**

### 109 **2.1 Study Design**

110 The study area is the city of Bologna, Italy. This is a highly polluted urban area of about 400,000  
111 inhabitants in northern Italy. In the period 2010-2012 the city-average annual mean number of  
112 exceedances of the daily PM<sub>10</sub> limit value (50 µg/m<sup>3</sup>) was 52.

113  
114 The main objective of the study was to compare exposure conditions of people living in residential  
115 settings with those in high traffic areas. The measurements at the two sites were conducted  
116 simultaneously indoors and outdoors at a residential as well as a traffic site. We selected indoor  
117 environments with the following characteristics: uninhabited, very similar in terms of volumes and  
118 building materials, and with very similar air exchange rates. The main goal was to assess the  
119 differences of population exposure to particles in relation to traffic without considering specific  
120 indoor characteristics and personal behaviours. We controlled the air exchange rates by installing in  
121 each indoor environment a mechanical system to force air to be exchanged between indoors and  
122 outdoors. The system consisted of an external fan connected to an air pipe (length = 1.2 m) carrying  
123 the air to the centre of the room (at a height of 2 m). Increased indoor air pressure caused the flow  
124 to exit the room through a grid. The fan in each room was set at a specific value related to the  
125 volume of the room in order to obtain an estimated 0.5 h<sup>-1</sup> air exchange rate in each room, a typical  
126 level for residential environments (Cattaneo et al., 2011). The air inflow was measured with a  
127 TESTO 417 Anemometer. There was concern that this experimental arrangement for providing a  
128 forced input of aerosol might lead to depletion due to passage through the fan and pipe. This  
129 possibility was tested by experiments in which the particle number size distribution was measured  
130 at the inlet to the fan and outlet to the pipe by an FMPS system with rapid switching between the

131 two sampling locations (upstream and downstream). The air inflow system was found to cause a  
132 minor loss of particles (additional information can be found in the Supplemental Information). This  
133 was considered a negligible source of error. The heating systems in the two indoor monitoring sites  
134 were kept always off.

135

136 The traffic site was located in a busy street which surrounds the historical centre of Bologna. This is  
137 one of the busiest streets of the entire municipal area with a traffic load of 31,000 vehicles (4–5%  
138 heavy duty vehicles) each working day. The building is located in a broad (20 m) two-way street  
139 canyon. The indoor monitoring site was on the ground floor in a two-storey building. The volume  
140 of the room was 55 m<sup>3</sup> with a ceiling height of 3.7 m. The floor was covered with marble. The  
141 ceilings and the walls were painted with acrylic paint.

142

143 The residential site was located in a low traffic area about 2 km from the historical centre of  
144 Bologna. The nearest street has a traffic volume of 6,000 vehicles per day. The measuring room was  
145 on the ground floor in a four-storey building. The volume of the room was 63 m<sup>3</sup> with a 3.7m  
146 ceiling height. The floor was covered with marble. The ceilings and the walls were painted with  
147 acrylic paint.

148

149 The outdoor PM<sub>2.5</sub> monitoring sites were located at 2 m above ground and for practical reasons at a  
150 distance of about 50 m from the indoor sites along the same streets. It is possible that the specific  
151 location of the outdoor PM<sub>2.5</sub> monitoring site could have produced a small reduction of the  
152 indoor/outdoor ratio for PM<sub>2.5</sub> and for some chemical components associated with primary  
153 emissions from traffic.

154

155 Three monitoring campaigns were conducted in the period February-June 2012. Each monitoring  
156 campaign lasted 15 days: 1<sup>st</sup> campaign from 22 February to 7 March, 2<sup>nd</sup> campaign from 16 to 30

157 April, and 3<sup>rd</sup> campaign from 28 May to 12 June. Filters were changed daily at each measurement  
158 site, and chemical speciation was performed sequentially every three days for metals, ions, and  
159 carbon (EC and OC). During the first two campaigns elemental and organic carbon were measured  
160 on an 8 hour basis in order to avoid an overload of the filters.

161

162

## 163 **2.2 Instrumentation and Monitoring Procedure**

164 Four identical low volume samplers (Skypost, TCR TECORA, Corsico – Mi) were operated to  
165 measure indoor and outdoor daily PM<sub>2.5</sub> concentrations at the two sites (flow rate 2.3 m<sup>3</sup>h<sup>-1</sup>). The  
166 samplers provide automatic filter changing after each 24-h period and are designed according to  
167 CEN standards. Each of these samplers consists of a PM<sub>2.5</sub> sampling inlet that is directly connected  
168 to a filter substrate and a regulated flow controller. Following completion of the sampling period,  
169 the PM<sub>2.5</sub> mass collected on the filter was determined gravimetrically. The filters were conditioned  
170 at 20°C and 50% relative humidity prior to weighing. Samples were collected on quartz fiber filters  
171 (Whatman, 47 mm diameter) and weighed following the procedure outlined in UNI EN 12341.

172

173 Agreements among the four instruments used in this study and some other identical instruments  
174 were checked in several intercomparison campaigns carried out in the years 2008-2012. Both  
175 correlation levels and test for differences for slope and intercept ( $1 \pm 2$  standard error (s.e.),  $0 \pm 2$   
176 s.e., respectively) of orthogonal regressions between co-located instruments were used as statistical  
177 indicators (EC, 2010). Determination coefficients were always higher than 0.972 (mean correlation  
178 0.985). Typical errors (standard deviation of the differences between samplers) were about 2 µg/m<sup>3</sup>  
179 and were quite similar among the various intercomparisons. The differences for slope (from unity)  
180 and intercepts (from zero) were usually not significant and not related to specific instruments.  
181 Consequently, no corrections were applied to PM<sub>2.5</sub> data.

182



183 PM<sub>2.5</sub> samples were analyzed for a various chemical species. In this paper we present the findings  
184 of the chemical species having more than 50% of contemporary data above the limit of  
185 quantification (LOQ) for indoor and outdoor samples at both sites. LOQs for chemical components  
186 were 0.028 µg/m<sup>3</sup> for iron (Fe), 0.04 µg/m<sup>3</sup> for ammonium (NH<sub>4</sub><sup>+</sup>), 0.04 µg/m<sup>3</sup> for potassium (K<sup>+</sup>),  
187 0.05 µg/m<sup>3</sup> for nitrates (NO<sub>3</sub><sup>-</sup>), 0.09 µg/m<sup>3</sup> for sulfates (SO<sub>4</sub><sup>2-</sup>), 2.1 µg/m<sup>3</sup> for daily Organic Carbon  
188 (OC), 0.3 µg/m<sup>3</sup> for daily Elemental Carbon (EC). OC and TC were quantified by means of  
189 thermal-optical transmittance (Sunset Laboratory Inc., USA) using the EUSAAR\_2 protocol.  
190 Inorganic ions were determined by extracting species in 10mL of ultrapure water. The extracts were  
191 filtered and analyzed by Ion Chromatography (Dionex ICS-1000 for anions and ICS-1100 for  
192 cations, Thermo Fischer Scientific Inc., USA). Iron was analyzed by Inductively Coupled Plasma –  
193 Mass Spectrometry (8800 ICP-MS, Agilent Technologies Inc., USA). Sample digestion was made  
194 with nitric acid and hydrogen peroxide in a microwave digestion apparatus, according to  
195 UNI14902:2005, with a recovery efficiency over 85%

196

197 Two Fast Mobility Particle Sizers (FMPS model 3091; TSI, Shoreview, MN, USA) were used to  
198 measure particle size distributions and to estimate UltraFine Particle (UFP) concentrations. The  
199 FMPS was developed based on electrical aerosol spectrometer technology from Tartu University  
200 (Tammet et al. 2002). The instrument consists of a particle charger column, a classification column,  
201 and a series of detection electrometers. After passing through the cyclone, the aerosol flow passes  
202 through a negative charger to prevent overcharging, and then a positive charger which applies a  
203 predictable charge on the sample using a corona unipolar diffusion charger. Small particles with  
204 high electrical mobility are repelled to the electrometers near the top of the column, and large  
205 particles with low electrical mobility are deflected further downstream. The particles transfer their  
206 charges to the electrometers generating currents that are inverted to produce a particle size  
207 distribution. The FMPS spectrometer measured the size and number concentration of particles from  
208 5.6 nm to 560 nm with 32 size bins every one second. Size bins below 13 nm were not included in

the analysis because of the amount of data below the detection limit and also because of questionable peaks in the size distribution observed in other studies. (Kaminski et al., 2013) (Jeong et al., 2009). UFP concentrations were obtained summing the number of particles detected in the channels below 100 nm. Hourly and daily data were calculated and used in the analyses. In the preliminary phase of the monitoring campaign we carried out a 1-week intercomparison between the two spectrometers using the same methodology applied for PM<sub>2.5</sub> samplers. We applied orthogonal linear regressions between data of each bin of the two instruments. Table 1S (Supplementary Information) shows the regression coefficients for each size bin. We found significant but small differences in the slopes and intercepts for the majority of size bins. Based on these findings we decided to apply bin-specific correction factors calculated during the intercomparison campaign to the data collected from one spectrometer during the field monitoring campaigns. The aim of this correction was to obtain an improvement of the comparability between the two FMPS. UFP number concentrations were calculated after the correction. Typical errors (standard deviation of the differences between UFP hourly data from the intercomparison campaign after the correction) were 320 part./cm<sup>3</sup>, and the determination coefficient was 0.989.

Nearly simultaneous indoor and outdoor size distributions were obtained with a switching system (Mod 11sc200, Pneumoidraulica Engineering S.r.l., Vicenza, Italy) which allowed for sampling from indoor and outdoor air, switching from one to the other within a time frame set by the user. A valve installed in the system could switch between sampling from the outdoor air, or from the indoor air. After the valve switched, there was a short time delay before the air from the sampled environment reached the instruments, which was the time the air travelled from the valve to the instruments. The system switched every 10 min between the indoor and outdoor measurements. In order to avoid the possibility of mixing of the outdoor and indoor air streams, the 2 min samples taken at the beginning of each 10 min period were deleted from the database.

235 Two digital thermo-hygrometers (Testo 175 H2, Testo AG, Lenzkirch, Deutschland) were used to  
236 measure temperature and relative humidity in the two indoor environments. Data were collected  
237 every 30 minutes.

238

### 239 **2.3 Analysis**

240 Summary statistics and paired t-test results have been calculated to investigate differences between  
241 series of measurements. Pearson correlation coefficients and regression analysis has been used to  
242 address linear relationships between data. We adopted an orthogonal regression approach (Fuller,  
243 1987) which is the most suitable when both dependent and independent variable are affected by  
244 errors and are not related by a causal relationship.

245 Quality control of PM<sub>2.5</sub> mass and chemical composition data was carried out based on residuals  
246 calculated by regression analysis between indoor and outdoor data. We identified as anomalous  
247 (not necessarily not valid) the data with residuals larger than three times the standard deviation of  
248 residuals.

249

250 For FMPS data quality control we used the following procedure: a) applying a log<sub>10</sub> function on  
251 the UFP minute data; b) stratifying data in time slots of three hours (0-3, 3-6 etc) and calculating the  
252 summary statistics for each slot and campaign; c) classifying data as anomalous if they were higher  
253 than the mean plus three times the standard deviation for the corresponding campaign and time slot.  
254 Then we averaged non-anomalous data on an hourly and daily basis.

255

256 Statistical data analysis was carried out using the R package (Version 3.0.1).

257

258

259

260

### 261 3. RESULTS AND DISCUSSION

#### 262 3.1 Measurement Campaigns and Meteorological Conditions

263 The meteorological conditions during the study periods are summarized in Table 2S  
264 (Supplementary Information). The sampling periods were quite representative of the typical annual  
265 variations in the area. The first campaign took place after an heavy snow event and was  
266 characterized by minimum temperatures similar to the typical values of the period but maximum  
267 temperature significantly higher than the climatological average values. In particular, in the second  
268 part of the first monitoring period maximum temperature reached 21.7°C, i.e. 4.2°C higher than the  
269 typical maximum temperature of the period. The second campaign was characterized by varying  
270 weather conditions with rainy and sunny days, and the third campaign was a typical early summer  
271 period. The area is characterized by low wind intensities and this was a common characteristic of  
272 the three monitoring campaigns (mean wind intensities from 1.9 m/s during the third campaign to  
273 2.6 m/s during the first campaign). Reasonably constant was also relative humidity which showed  
274 very similar mean values (55.2, 53.8, 50.4% for the three campaigns) but large day to day  
275 variations. Precipitation events were rare and small for all the sampling periods. In particular during  
276 the first campaign we had only three rainy days with 2 mm mean precipitation.

277

278 The temperatures measured at the two indoor monitoring sites showed very similar values and  
279 temporal patterns. Seasonal differences were clearly reduced compared to the outdoor values  
280 especially due to higher minimum temperatures. On the contrary, we found larger seasonal  
281 variations in indoor compared to outdoor RH values although RH indoor values were always below  
282 50%.

283

284

285

## 286    **3.2        Measurements of Indoor and Outdoor Particles**

287    It is important to recognise that the design of this experiment is different from most earlier work on  
288    indoor and outdoor particle measurements. Most earlier studies have depended upon natural  
289    ventilation processes involving exchange of air through cracks around doors and windows or  
290    through open windows which lead to air exchange. In these experiments, air exchange was forced  
291    by a fan driving air into the room and consequently the processes of particle loss will be subject to  
292    some differences. In the absence of indoor sources, indoor particle concentrations are generally  
293    found to be lower than those outdoors due to particle loss on surfaces during the infiltration of air  
294    and due to loss on internal surfaces within the building. In these experiments, the first loss  
295    mechanism is insignificant as the air introduction method caused only very small changes to particle  
296    concentrations and hence the reductions in airborne concentrations are due almost solely to  
297    deposition to surfaces. The removal of air by the FMPS and filtration of particles by the PM<sub>2.5</sub>  
298    sampler are at a rate far smaller than the air exchange for the room and consequently have only a  
299    modest influence upon the measured indoor concentrations.

300

301    In their review article, Chen and Zhao (2011) define both an Infiltration Factor which represents the  
302    equilibrium fraction of particles which penetrates indoors and remains suspended, and a Penetration  
303    Factor which describes the penetration efficiency of particles through the building envelope. In our  
304    study design, the Penetration Factor is 1.0 (100%), and we measure an Infiltration Factor.

305

### 306    **3.2.1        Comparison of the sites**

#### 307    **3.2.1.1    Indoor and outdoor PM<sub>2.5</sub> mass**

308    Indoor and outdoor PM<sub>2.5</sub> concentrations during the three monitoring campaigns are shown in  
309    Figure 1 (upper panel) and Table 1. No PM<sub>2.5</sub> data was identified as anomalous and removed from  
310    the database. Outdoor concentrations in the first campaign were about three times higher compared

311 to the other two monitoring periods. Very small variations (less than 10%) were found between the  
312 second and third campaign. Much higher concentrations during the winter season are typical of the  
313 area (Bigi et al., 2012).

314  
315 We found very small and non-significant differences in  $PM_{2.5}$  outdoor concentrations between the  
316 sites (see Figure 2). Average  $PM_{2.5}$  concentrations at the traffic site were about 6% higher than at  
317 the residential site. The highest values for daily mean  $PM_{2.5}$  concentrations were  $72 \mu g/m^3$  for the  
318 traffic site and  $70 \mu g/m^3$  for the residential site. The  $PM_{2.5}$  spatial variability found in our study was  
319 a little lower than the mean within-city variability reported in the ESCAPE study, a very large  
320 epidemiological study in Europe which included monitoring campaigns on air pollution spatial  
321 variability in urban areas (Eefftens et al., 2012). In that study the mean ratio between traffic sites  
322 and urban background sites was 1.14, with a quite broad range of values (0.96 – 1.30). However, it  
323 should be taken into account that we compared a traffic with a residential site in a low traffic area,  
324 whilst background sites in many other studies have been placed in parks. In fact, our aim was to  
325 assess the variability of  $PM_{2.5}$  concentrations between areas where people live. In addition, a  
326 reduced relative spatial variability of  $PM_{2.5}$  could be explained also by the higher background  
327 contribution of secondary particulate matter to the total  $PM_{2.5}$  mass in this area (Perrino et al.,  
328 2013).

329  
330 Indoor/outdoor (I/O) ratios of  $PM_{2.5}$  were close to 0.4 during the first campaign at both monitoring  
331 sites. The I/O ratio increased in the subsequent campaigns with mean values equal to 0.9 at the  
332 traffic site and 0.7 at the residential site for the second and third campaign. This range of I/O ratios  
333 was in good agreement with previous studies on indoor settings (Chen and Zhao, 2011). Inter-  
334 campaign variations of  $PM_{2.5}$  in the indoor sites were lower than outdoors. The ratios between mean  
335 indoor concentrations during the first campaign and the other two were 1.6 and 1.3 for the traffic  
336 site and 2.3 and 1.8 for the residential site. The major difference in I/O ratio between the first

337 campaign and the latter two seems most likely related to the aerosol composition. The outdoor  
338 nitrate content was much higher in the cooler first campaign, leading to a much reduced I/O ratios  
339 (Figure 3). The higher I/O ratios observed at the traffic site in the second and third campaign seems  
340 most likely related to the higher I/O ratio for elemental and organic carbon and iron (Figure 3)  
341 which were the (traffic-related) constituents showing the largest difference between the sites.

342  
343 Values of Infiltration Factor for  $PM_{2.5}$  reviewed by Chen and Zhao (2011) range from around 0.35  
344 to 0.82. Penetration factors in the size range of 0.1-2.5  $\mu m$ , in which most  $PM_{2.5}$  mass resides are  
345 typically in the range of 0.75-1.0, with many measured values close to 1.0. Chen and Zhao (2011)  
346 highlight the anomalous behaviour of reactive particles such as nitrates. If a Penetration Factor of  
347 0.9 is applied to the above range of Infiltration Factors (0.35 to 0.82), it yields adjusted values of  
348 0.39 to 0.91 which should be, and are broadly equivalent to the I/O values determined in our study.  
349 The only divergences appear to be due to semi-volatile nitrates which lead to lower values of I/O  
350 ratio. The more recent review of Diapouli et al. (2013) also summaries results for the penetration  
351 efficiency and infiltration factor for  $PM_{2.5}$ . The former ranges from 0.54-1.0, with the majority of  
352 data in the 0.8-1.0 range. The infiltration factor lies between 0.4-0.85 in the various studies  
353 reviewed, which is very consistent with that reported above, no doubt because both reviews include  
354 many studies in common.

355  
356 Figure 2 shows the scatter plots and the correlation coefficients calculated among the measurement  
357 sites. We found a very high level of correlation between outdoor  $PM_{2.5}$  concentrations at the two  
358 sites. Indoor  $PM_{2.5}$  concentrations were highly correlated as well. Pearson coefficients were equal to  
359 0.97 for the outdoor correlations and 0.88 for the indoor correlations. Somewhat lower correlations  
360 were found between indoor and outdoor concentrations. I/O correlation coefficients at the traffic  
361 and the residential site were equal to 0.75 and 0.82, respectively. The latter coefficients are similar

362 to the highest values found in other studies (e.g. Hanninen et al., 2004) and this is probably related  
363 to the absence of indoor sources.

#### 364 **3.2.1.2 Indoor and outdoor UFP number concentrations**

365 Based on the procedure outlined in the methods section, 0.83% and 1.33% of minute data in the  
366 residential and traffic site, respectively were classified as outliers and removed from the database.  
367 The completeness of hourly data at the outdoor traffic site, indoor traffic site, outdoor residential  
368 site, indoor residential site was 100%, 98%, 86% and 73%, respectively. Completeness of data at  
369 the residential site was lower because the switching unit had problems during the first campaign and  
370 nighttime indoor data at the residential site were not available.

371  
372 Figure 1 (lower panel) and Table 1 give an overview of the ultrafine particle concentrations during  
373 the monitoring campaigns. Outdoor concentrations at the traffic site were much higher than at the  
374 residential site. Mean outdoor UFP concentrations measured at the traffic site during the three  
375 campaigns were 3.4, 3.2 and 1.7 times higher than at the residential site. The highest hourly value at  
376 the traffic site was 129,400/cm<sup>3</sup> while the highest value in the residential site was 37,790/cm<sup>3</sup>.  
377 These findings were in good agreement with the findings of studies carried out in Los Angeles  
378 (Moore et al., 2009) and in Spain (Rivera et al., 2012). Similar results were also found in another  
379 study in Athens (Diapouli et al., 2011) showing a spatial variability ranging from ratios of 1.8 to 2.6  
380 depending on the season. Significantly lower gradients were found in a study of four major  
381 European cities (Puustinen et al., 2007), but indoor sources were present.

382  
383 The indoor concentrations of UFP at the residential site varied over a relatively small range  
384 compared to the larger day to day variations evident at the traffic site (Figure 2 and Figure 4). The  
385 ratio between indoor UFP concentrations at the two sites varied between the three campaigns (Table  
386 1) with UFP levels at the traffic site approximately 2-4 times those of the residential site.

387



388 As with  $PM_{2.5}$ , the I/O ratio for UFP increased at the traffic site from 0.38 in the first campaign to  
389 0.69 in the third campaign. On the contrary, the I/O ratio at the residential site remained more  
390 constant at around 0.5. Diapouli et al. (2011) found an I/O ratio for particles in the 10-400 nm size  
391 range equal to 0.6 in the 0.5-1 range of AER while I/O ratios between 0.3-0.4 were found in Erfurt  
392 (Germany) (Cyrys et al., 2004) and in other major European urban areas (Hoek et al., 2008b)

393  
394 Ultrafine particles show marked increases between 7-9 a.m. and 7-8 p.m. (Figure 5) in  
395 correspondence with the rush hours. These peaks were significantly higher at the traffic site. The  
396 maximum concentrations in the morning were reached at 8 a.m. at the residential site and at 9 a.m..  
397 at the traffic site. During the afternoon the maximum was reached at 9 p.m. at the residential site  
398 and at 7 p.m. at the traffic site during the first campaign. In the second campaign the afternoon  
399 peaks were shifted one hour later. Morning peaks were typically higher than the late afternoon  
400 peaks. The differences in UFP concentrations between indoor levels at the two sites were quite  
401 constant during the day for all campaigns. The differences in concentrations between indoor levels  
402 decreased slowly during the night leading to almost identical indoor concentrations at the two sites  
403 at around 5 a.m.. These results were in good agreement to those reported by Lianou et al. (2011).

404

405 The Pearson correlation coefficient between daily outdoor UFP concentrations at the two sites was  
406 equal to 0.89, significantly higher than those reported in other studies of particle number  
407 concentrations (Puustinen et al., 2007). Correlations between indoor UFP concentrations were much  
408 lower ( $R=0.42$ ). Very similar correlation coefficients were found between indoor and outdoor UFP  
409 concentrations at the traffic and residential sites ( $R= 0.57$  and  $0.63$  respectively). A broad range of  
410 correlations between I/O daily data was found by Hoek et al. (2008b) with values ranging from 0.41  
411 in Helsinki to 0.80 in Athens. The correlation coefficient between hourly outdoor concentrations  
412 was equal to 0.71. Slightly higher correlations between indoor hourly data were found than for the  
413 daily data, with I/O hourly correlation coefficients almost equal to 0.60 for both sites.

414 With the exception of the third campaign, I/O ratios were markedly higher at the residential site  
415 than the traffic site (Table 1). This seems most probably related to more efficient loss of the traffic-  
416 generated ultrafine particles which predominate at the traffic site, as seen in Figure 6. The higher  
417 temperatures in the third campaign probably minimised the contribution of this aerosol component  
418 due to its semi-volatile nature (Fujitani et al., 2012) as reflected in the lower outdoor particle  
419 number counts in this campaign (Table 1).

420

### 421 **3.2.1.3 Indoor and outdoor size distribution**

422 Figure 6 shows the mean indoor and outdoor size distributions at the two sites. Multimodal  
423 distributions with sharp peaks at about 30 nm in the outdoor concentrations were found at the traffic  
424 site. The same peak was present also at the other indoor and outdoor monitoring sites though much  
425 less pronounced. A second peak was present at about 80-100 nm. This is typical of heavily  
426 trafficked sites, with the modes arising from the semi-volatile nucleation particles and solid  
427 graphitic particles respectively (Harrison et al., 2011). The plateau in the indoor distribution of the  
428 traffic site may be the joint effect of the coexistence of two modes, an Aitken mode peak at about  
429 40-50 nm and an accumulation mode peak at 100-200 nm. The two principal peaks were evident for  
430 all hours during the day and all monitoring campaigns although their relative weight was highly  
431 variable, especially during the day. Figure 7 shows the huge increase of the 30 nm peak during the  
432 rush hours. The highest particle number concentrations were found at 9 a.m. (i.e. between 8 a.m.  
433 and 9 a.m). The subsequent hours showed a decrease of this peak with a further increase in the late  
434 afternoon (5 p.m. to 8 p.m.). The differences between morning and late-afternoon of particle  
435 number concentration in the 30 nm size range were more evident during the first campaign and  
436 decreased in the following campaigns. Similar patterns were seen at the residential site with a much  
437 less pronounced peak in the 30 nm size range. Indoor particle concentrations were lower compared  
438 to outdoor concentrations for both sites and all size bins. Very similar particle size distributions

439 were found after midnight at both indoor and outdoor sites. Indoor size distributions were similar to  
440 the findings of Hussein et al. (2004) with an increase of the nucleation mode associated with rush  
441 hours. Minor intra-day variations were found for particles in the accumulation mode. Much lower  
442 relative weight of the nucleation mode compared to the accumulation mode was found indoors  
443 compared to the outdoor size distributions, as reported in previous studies (Hussein et al., 2004;  
444 Diapouli et al., 2011). The comparison of the indoor size distributions between sites showed similar  
445 trends but relevant absolute differences. Differences in the nucleation mode were much reduced  
446 compared to outdoor distributions. This was probably due to the importance of size-dependent  
447 removal mechanisms that show a maximum in the lower and upper part of the spectrum. (Riley et  
448 al., 2002)

449

450 Diapouli et al. (2013) review data for ultrafine particles from three studies for penetration  
451 efficiency, showing values from 0.47-0.80. For infiltration factor, two studies give values close to  
452 0.60 (Diapouli et al., 2013). While caution is needed in making comparisons of studies due to the  
453 high size-dependence of ultrafine particle losses, these values imply I/O ratios similar to the I/O  
454 ratios in Table 1 for UFP, which range from 0.38-0.69. The application of the highest penetration  
455 efficiency of 0.80 to an infiltration factor of 0.60 (both figures from Diapouli et al., 2013) suggests  
456 an I/O ratio equivalent to that in our experiment of 0.75.

457

458 The presence of a bi (or tri)-modal distribution was also shown in previous studies (Morawska et  
459 al., 2008; Hussein et al., 2005) and is in line with knowledge of particle emissions and  
460 transformation. The 30 nm mode is due to the combination of freshly nucleated particles formed as  
461 the exhaust gases are diluted with ambient air and particles directly emitted by vehicles (Charron et  
462 al., 2003). Particles emitted from diesel engines are in the size range 20–130 nm and from petrol  
463 engines in the range 20–60 nm (Ristovski et al., 2006). Emission factors for petrol (gasoline) cars  
464 are much lower than for diesel (Beddows and Harrison, 2008), although petrol vehicles during

465 acceleration show particle number emissions close to those observed from diesel vehicles (Graskow  
466 et al., 1998). Nucleation mode particles are associated with the hot exhaust gases expelled from the  
467 tailpipe of a vehicle. These gases cool and condense to form large numbers of very small particles  
468 in the air (Shi and Harrison, 1999). On-road dilution of the exhaust plume is very important in the  
469 generation of particles in the exhaust plume. These nucleation processes are favoured by low  
470 ambient temperatures and high relative humidity (Charron et al., 2003). In addition, the gaseous  
471 precursors condense or adsorb on to the surface of carbon particles in the accumulation mode. If the  
472 concentration of carbon particles is low, the gases will nucleate homogeneously, giving rise to large  
473 concentrations of semi-volatile nanoparticles.

474  
475 The differing meteorological conditions between the first campaign and the other campaigns can  
476 explain the differing indoor/outdoor ratios. The cooler atmospheric conditions of the first campaign  
477 would tend to increase the semi-volatile nucleation mode particles relative to the coarser graphitic  
478 mode particles in the traffic aerosol. Upon entry into the building, not only would the nucleation  
479 mode fraction show a higher deposition velocity than the coarser graphitic mode (Riley et al.,  
480 2002), it would be subject to evaporation at the higher indoor temperatures (Dall'Osto et al., 2011)  
481 and the hydrocarbon vapours released would tend to adsorb to indoor surfaces (Weschler and  
482 Nazaroff (2008) and settled indoor dusts (Weschler and Nazaroff, 2010). Such processes would  
483 contribute to a relatively rapid loss of the nucleation mode of particles hence explaining both the  
484 changes in size distribution seen in Figure 5 and the far higher outdoor/indoor ratios seen at the  
485 trafficked site in the first campaign (Table 1). Semi-volatile components of the PM<sub>2.5</sub> might also  
486 show lower I/O ratios in cold weather due to enhanced volatilisation in the warmer indoor  
487 environment.

488

489 It is clear from Figures 6 and 7 that a large proportion of the sub-200 nm particles have been lost  
490 between the traffic and residential sites, as a result of dispersion processes, and of evaporation for

491 the smaller sized particles. However, as these particles are also lost with high efficiency in the  
492 indoor environment, the mean indoor size distributions differ little between the sites, although a  
493 concentration difference remains (Figure 6).

494

#### 495 **3.2.1.4 Indoor and outdoor chemical composition of PM<sub>2.5</sub>**

496 All concentrations of chemical components had more than 75% of values above LOQ with the  
497 exceptions of residential indoor iron (73% of data above LOQ), traffic indoor potassium (60%),  
498 residential indoor potassium (53%). No data on chemical composition of PM<sub>2.5</sub> was identified as  
499 anomalous and removed from the database. OC was found to be the largest contributor to outdoor  
500 PM<sub>2.5</sub> mass at both sites followed by nitrates, elemental carbon and sulfates. The contribution of  
501 OC to indoor PM<sub>2.5</sub> was even larger followed by elemental carbon and sulfates. Mean  
502 concentrations appear in Figure 3.

503

504 Significant differences (paired t-test, significance level = 0.05) between traffic outdoor and  
505 residential outdoor data were found for iron, elemental carbon and total carbon: 222 vs 135 ng/m<sup>3</sup>  
506 for iron, 2.9 vs 1.8 µg/m<sup>3</sup> for elemental carbon. Minor and non-significant differences were found  
507 for the other chemical components. Very similar spatial gradients were found for indoor data. A  
508 large impact of traffic proximity on iron and carbonaceous species has been reported in several  
509 studies. Iron has been found an elemental marker for both exhaust and non-exhaust emissions (Pant  
510 and Harrison, 2013) while carbonaceous particles have been related mainly to exhaust emissions.

511

512 Indoor concentrations were lower than outdoor for all chemical species and both sites with the only  
513 exceptions of elemental carbon at the traffic site. The higher concentrations of elemental carbon  
514 found indoors at the traffic site may have been caused by the specific location of the outdoor  
515 measurement site. As already mentioned in Section 2.1, the outdoor monitoring site was some 50 m  
516 away from the indoor site in a location which was less influenced by the canyon effect. Very large

517 differences were found especially for nitrates, ammonium, potassium and sulfates. The lowest I/O  
518 ratio was observed for nitrates. Average nitrate concentrations at the two outdoor measurements  
519 sites were 4.6 and 4.7  $\mu\text{g}/\text{m}^3$  while indoor concentrations were equal to 0.3  $\mu\text{g}/\text{m}^3$ . I/O ratios for  
520 ammonium and sulfates were 0.3 and 0.6, respectively. The outdoor and indoor EC/TC ratios were  
521 respectively 0.35 and 0.4 at the traffic site and 0.24 and 0.13 in the residential site. These values are  
522 similar to those reported for outdoor urban air data by Naser et al. (2008).

523  
524 Table 2 shows the Pearson correlation coefficients calculated among the chemical components data.  
525 Outdoor data at the two sampling sites were highly correlated (correlation coefficients always  
526 greater than 0.9). High correlation levels between within-city outdoor concentrations of organic  
527 carbon, elemental carbon, ammonium, nitrates and sulfates was found by Bell et al. (2011). Similar  
528 results were found for organic carbon and elemental carbon by Naser et al. (2008). High  
529 correlations were found also between indoor data with the exception of iron ( $R=0.33$ ). Correlation  
530 levels were also generally very high in respect of I/O data. Low values were found only for  
531 ammonium ( $R=0.24$ ) and iron ( $R=0.38$ ) at the traffic site and ammonium ( $R=0.45$ ) at the residential  
532 site. High correlations between indoor and outdoor concentrations of organic carbon and elemental  
533 carbon were also found by Sawant et al. (2004) in several schools in California. The I/O  
534 correlations for ammonium found by Sawant et al. (2004) showed large variability in the different  
535 schools. Particles of outdoor origin can undergo substantial changes and may be lost to building  
536 walls during indoor penetration. A study investigating the transformation of ambient ammonium  
537 nitrate aerosols in indoor environments has shown that measured indoor concentrations were  
538 considerably lower than the values predicted based only on penetration and deposition losses  
539 (Lunden et al., 2003). This is due to the semi-volatility of ammonium nitrate, leading to loss as  
540 nitric acid vapour and ammonia which attach to indoor surfaces. Varying ratios of volatile  
541 ammonium nitrate to involatile ammonium sulphate will lead to varying indoor-outdoor ratios of  
542 ammonium and hence the weaker correlation.

543 The only major inter-site differences in behaviour appearing in Figure 3 are for iron and elemental  
544 carbon. Both are primary emissions from road traffic and the apparent behaviour at the traffic site  
545 is probably anomalous because of the spatial separation of the indoor and outdoor samplers referred  
546 to above.

547

### 548 **3.3 Strengths and Weaknesses of the Study**

549 A major strength of the study was the contemporary measurements of a number of particle metrics  
550 and characteristics in indoor and outdoor environments with very different characteristics in relation  
551 to traffic sources. The absence of indoor sources and the attention devoted to make air exchange  
552 rates as similar as possible should reduce to a minimum the noise due to personal behavior and  
553 specific indoor characteristics. Thus concentration gradients between sites should be almost solely  
554 due to proximity to traffic sources.

555

556 A weakness was related to the use of a very simple system to impose an air exchange rate in the two  
557 environments. Direct measurements of the air exchange rates were not made. However, it should be  
558 taken into account that air exchange rates are almost always measured at only one or a few points in  
559 time during a monitoring campaign. Therefore this is a general weakness of this type of study  
560 because air exchange rates vary significantly in time in relation to outdoor conditions and several  
561 other factors. Even if AER had been measured directly we would not have had much greater  
562 guarantee on the exact level of AER in the two indoor environment during the 45 day measurement  
563 period. Moreover, the main goal was to have similar AER in the two indoor environments and this  
564 goal should have been achieved using two identical systems for ventilation. Our experimental  
565 system should facilitate measurements under different controlled ventilation conditions.  
566 Additionally, it has the benefit of allowing evaluation of indoor deposition processes independently  
567 of losses during infiltration through the building shell.

568

#### 569 4. SUMMARY AND CONCLUSIONS

570 This study addressed the issue of the difference in exposure to particles in relation to proximity to  
571 traffic within an urban area. In particular, we analysed indoor and outdoor PM<sub>2.5</sub> mass and chemical  
572 composition, size distribution and particle number concentrations in a heavy traffic site compared to  
573 a residential area.

574  
575 Large spatial variability in the concentrations of UFP, iron and elemental carbon was found both  
576 indoors and outdoors. Concentrations of UFP were much higher at the traffic site. Mean indoor  
577 concentrations at the traffic site were higher than outdoor concentrations at the residential site.  
578 Indoor variability was higher than outdoors for iron and elemental carbon. Significant differences  
579 were also found for the shape of particle size distributions for outdoor particles while indoor  
580 particles showed very similar distributions. Indoor concentrations were much lower than outdoor  
581 for PM<sub>2.5</sub> mass and UFP, especially when outdoor concentrations were high and air temperatures  
582 low. Taking into account the chemical components, deposition to the building surfaces was  
583 protective especially for nitrates, ammonium, potassium, and sulfates. Both indoor and outdoor  
584 PM<sub>2.5</sub> concentrations were highly correlated while indoor UFP correlations were much lower than  
585 outdoor. The temporal trends of all chemical species at the two indoor sites were highly correlated  
586 with the exception of iron.

587  
588 Our findings represent a contribution to understanding the appropriate particle metrics and data to  
589 be collected in epidemiological studies of the health effects of air pollution. In particular, our  
590 analyses showed that fixed site monitoring stations represent quite well the temporal trend of  
591 population exposure for PM<sub>2.5</sub> together with its chemical components. Although indoor exposure  
592 could be significantly lower than outdoor, they are spatially very well correlated considering both  
593 indoor and outdoor concentrations. Some caution should be adopted for iron which showed high  
594 correlation between outdoor data but low correlation between indoor data. UFP concentrations



595 showed lower correlations compared to  $PM_{2.5}$ , in particular between indoor concentrations.  
596 Therefore fixed site stations could be less representative of the temporal trends of population  
597 exposure. With regards to size distribution, an important decrease of the relative concentration of  
598 the nucleation mode relative to the accumulation mode in the indoor air compared to the outdoor  
599 was found at the traffic site.

600  
601 With regards to the epidemiological studies aiming at assessing the health impact of proximity to  
602 traffic, we observed that largest gradients in exposure were found for UFP, iron and elemental  
603 carbon. Tiny and insignificant differences were found for  $PM_{2.5}$  and the other chemical components.  
604 Indoor spatial gradients generally reflected outdoor gradients quite closely.

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865 **TABLE LEGENDS**

866

867 **Table 1** Summary statistics of PM<sub>2.5</sub> mass ( $\mu\text{g m}^{-3}$ ) and UFP number ( $\text{cm}^{-3}$ ) during the  
868 monitoring campaigns.

869

870 **Table 2** Pearson correlation coefficients for different chemical species.

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874 **FIGURE LEGENDS**

875

876 **Figure 1** Average value and standard error of PM<sub>2.5</sub> (upper panel) and UFP (lower panel) during  
877 the three monitoring campaign.

878

879 **Figure 2** Scatter plot for daily PM<sub>2.5</sub> mass and UFP number.

880

881 **Figure 3** Scatter plot for hourly UFP number.

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883 **Figure 4** Daily temporal trend of UFP number during the three monitoring campaigns.

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885 **Figure 5** Mean particle size distribution of hourly data.

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887 **Figure 6** Mean particle size distribution of hourly data at specific time of the day.

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889 **Figure 7** Average value and standard errors of the chemical species.

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891

Table 1. Summary statistics of PM<sub>2.5</sub> mass ( $\mu\text{g m}^{-3}$ ) and UFP number ( $\text{cm}^{-3}$ ) during the monitoring campaigns.

	Traffic					Residential				
	Number of valid data	Outdoor mean (min - max)	Number of valid data	Indoor mean (min - max)	I/O mean	Number of valid data	Outdoor mean (min - max)	Number of valid data	Indoor mean (min - max)	I/O mean
<b>PM2.5 (24-hour data)</b>										
All data	45	22.5 (5.1 - 72.0)	45	13.0 (5.3 - 27.0)	0.74	45	21.3 (3.9 - 70.0)	45	10.0 (3.1 - 31.0)	0.59
1 <sup>st</sup> campaign	15	40.7 (20.0 - 72.0)	15	16.3 (11.0 - 27.0)	0.42	15	38.4 (18.0 - 70.0)	15	15.1 (8.0 - 31.0)	0.40
2 <sup>nd</sup> campaign	15	12.9 (5.1 - 27.6)	15	10.2 (5.3 - 15.2)	0.88	15	13.0 (3.9 - 28.7)	15	6.6 (3.1 - 10.8)	0.70
3 <sup>rd</sup> campaign	15	14.0 (7.7 - 18.5)	15	12.6 (8.4 - 18.1)	0.92	15	12.5 (6.4 - 16.1)	15	8.4 (6.0 - 13.7)	0.69
<b>UFP (1-hour data)</b>										
All data	1,075	24,006 (2,193 - 129,386)	1,063	8,641 (1,418 - 21,933)	0.48	934	6,810 (1,446 - 37,790)	785	2,836 (396 - 13,375)	0.52
1 <sup>st</sup> campaign	360	31,042 (3,973 - 129,386)	360	9,117 (2,568 - 21,933)	0.38	292	10,148 (3,642 - 37,790)	146	4,885 (2,969 - 13,375)	0.54
2 <sup>nd</sup> campaign	357	25,752 (2,193 - 108,588)	353	7,939 (1,418 - 20,407)	0.38	282	6,024 (2,156 - 22,191)	283	2,439 (1,315 - 6,088)	0.47
3 <sup>rd</sup> campaign	358	15,189 (2,552 - 60,773)	350	8,859 (2,363 - 20,952)	0.69	360	4,718 (1,446 - 13,988)	356	2,311 (396 - 8,287)	0.54

Sampling periods: 1<sup>st</sup> campaign from 22 February 2012 to 7 March 2012; 2<sup>nd</sup> campaign from 16 to 30 April 2012; 3<sup>rd</sup> campaign from 28 May to 12 June



Table 2. Pearson correlation coefficients for different chemical species.

	Traffic indoor vs traffic outdoor			Residential indoor vs residential outdoor			Traffic outdoor vs residential outdoor			Traffic indoor vs residential indoor		
	R	Slope	Intercept ( $\mu\text{g}/\text{m}^3$ )	R	Slope	Intercept ( $\mu\text{g}/\text{m}^3$ )	R	Slope	Intercept ( $\mu\text{g}/\text{m}^3$ )	R	Slope	Intercept ( $\mu\text{g}/\text{m}^3$ )
Iron (Fe)	0.38	0.64	0.066	0.87	0.29	0.014	0.9	1.62	0.003	0.33	11.61	-0.397
Ammonium ( $\text{NH}_4$ )	0.23	0.08	0.48	0.42	0.12	0.35	0.99	0.97	0.06	0.95	1.09	-0.01
Nitrates ( $\text{NO}_3$ )	0.86	0.03	0.17	0.81	0.04	0.16	1	0.97	0.04	0.93	0.69	0.07
Sulfates ( $\text{SO}_4$ )	0.99	0.72	-0.22	0.94	0.69	-0.33	0.97	1.12	-0.35	0.96	1.15	-0.07
Organic carbon (OC)	0.91	0.52	2.42	0.88	0.58	2.21	0.99	0.9	0.65	0.97	0.8	1.02
Elemental carbon (EC)	0.77	0.74	1.2	0.94	0.48	0.05	0.98	0.83	1.48	0.68	1.51	2.03
Total carbon (TC)	0.93	0.55	3.91	0.92	0.55	2.27	0.99	0.89	2.11	0.91	0.88	3.12

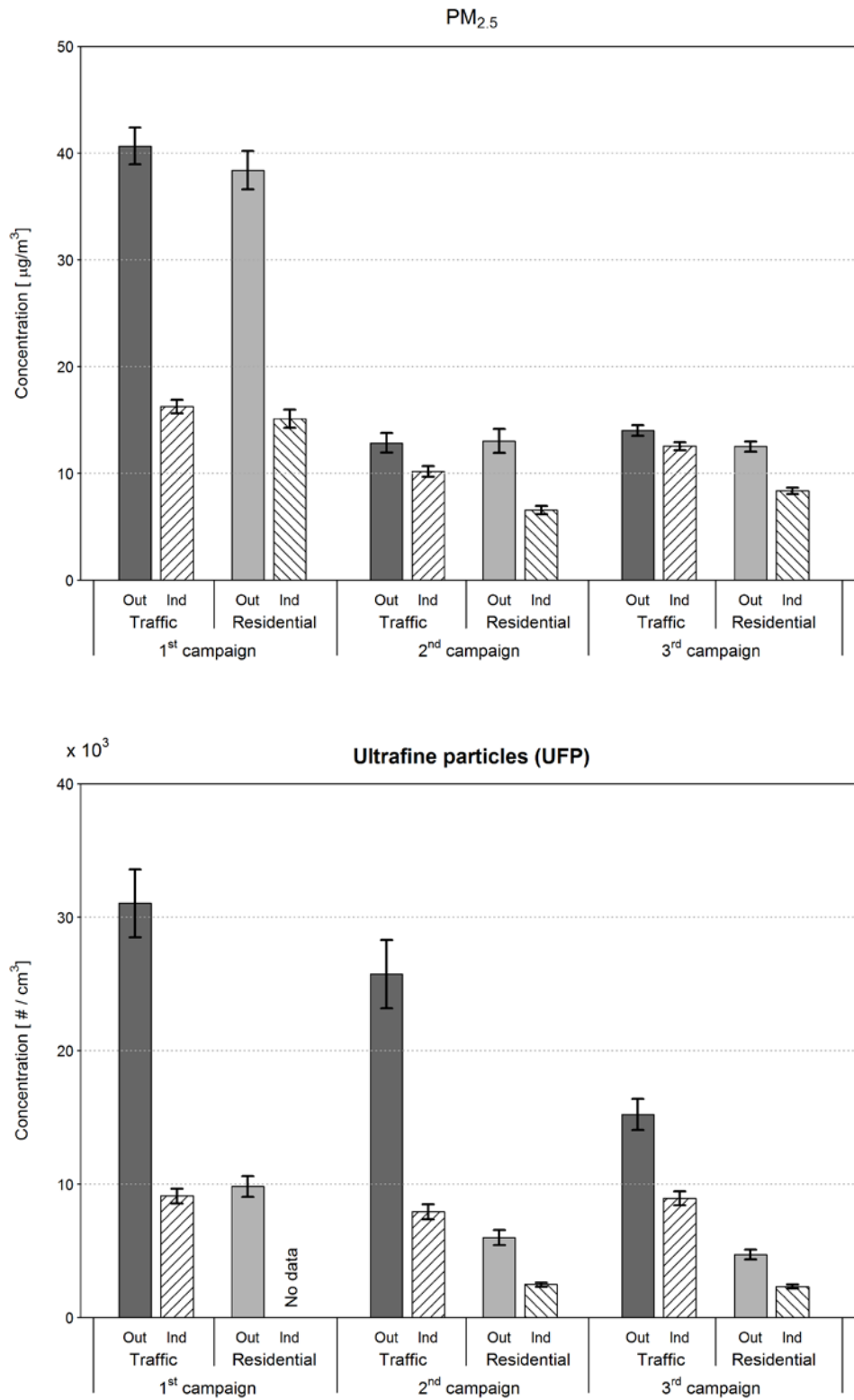


Figure 1. Average value and standard error of  $PM_{2.5}$  (upper panel) and UFP (lower panel) during the three monitoring campaign.

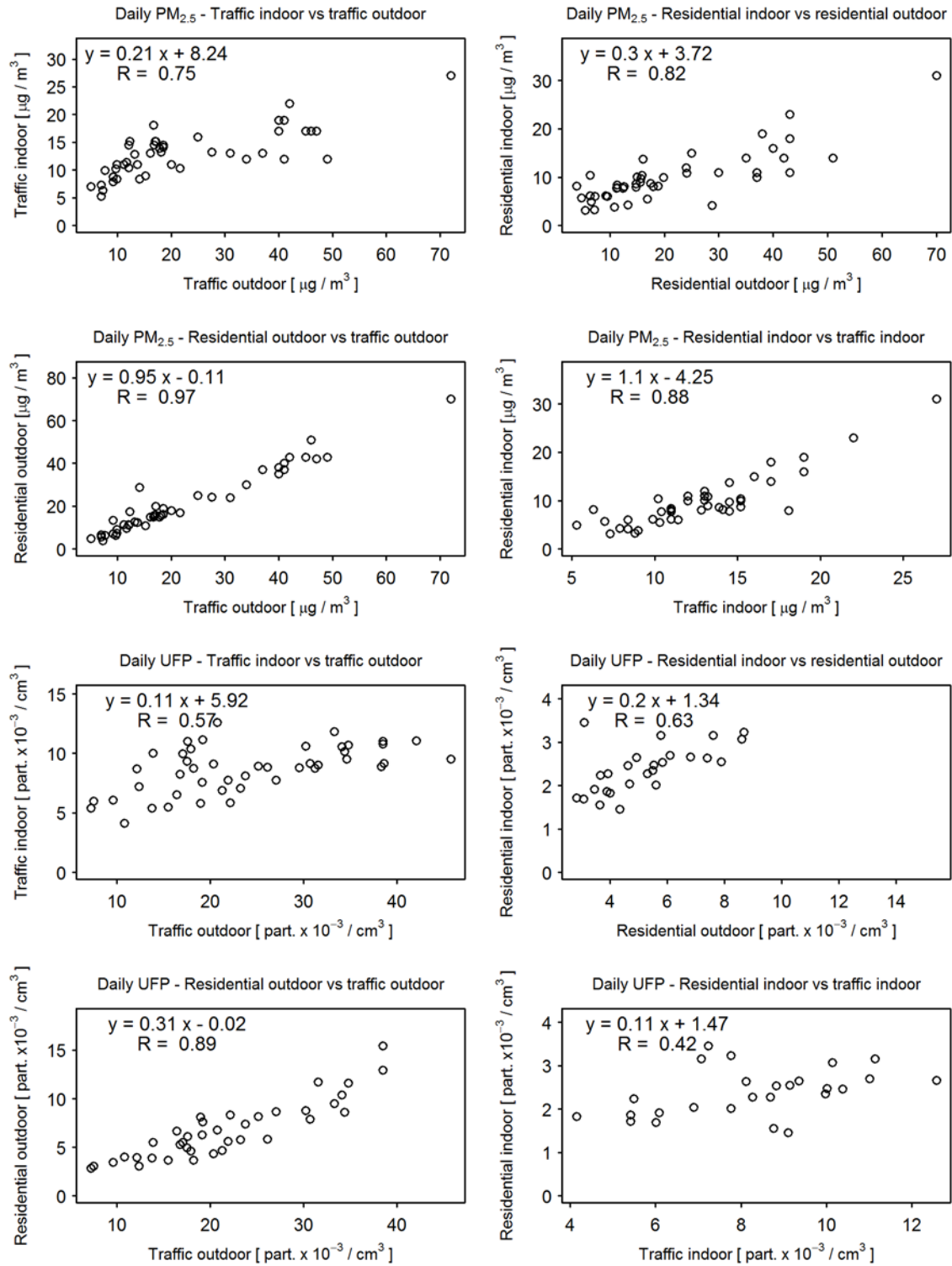


Figure 2. Scatter plot for daily PM<sub>2.5</sub> mass and UFP number.

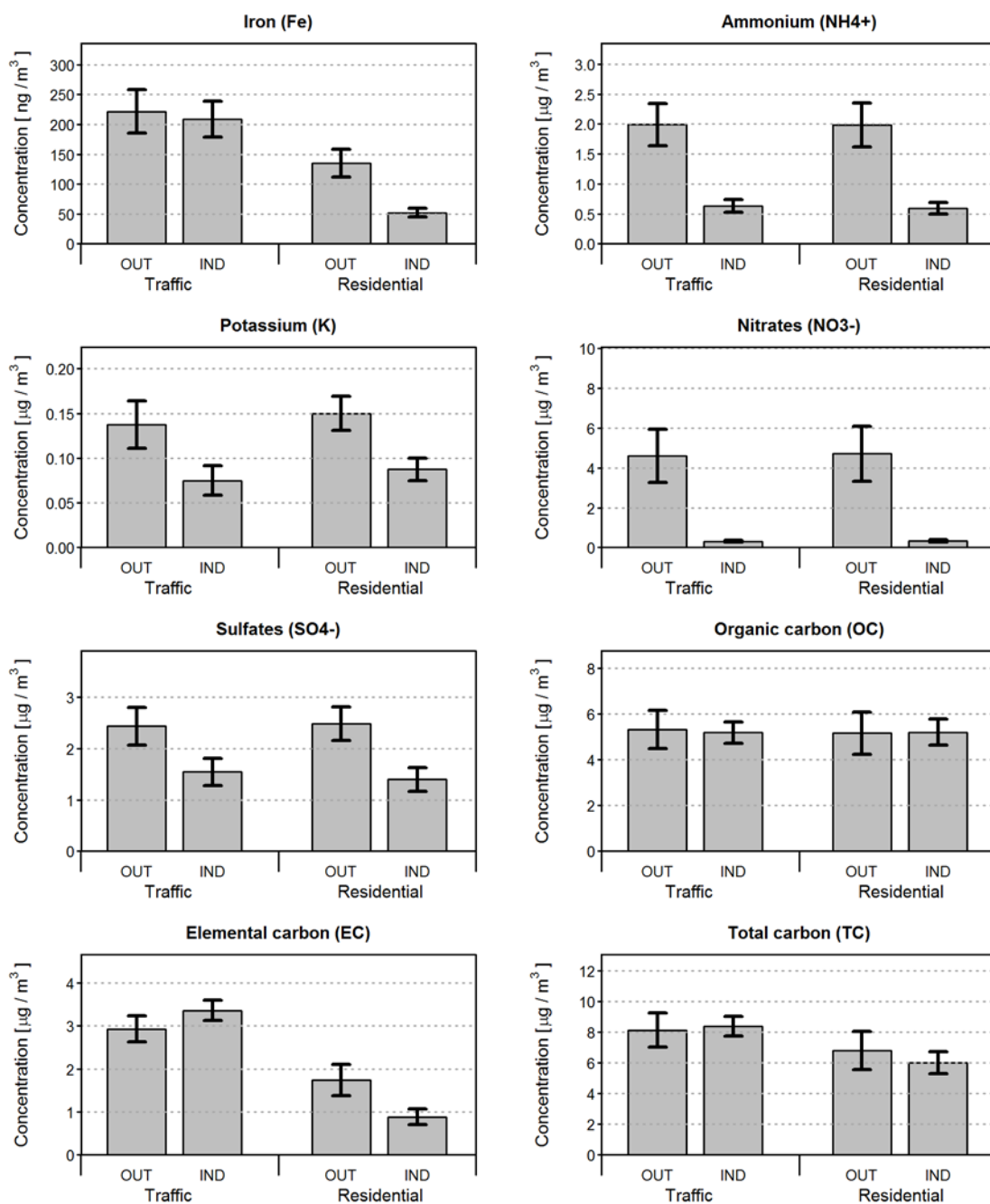


Figure 3. Average value and standard errors of the chemical species.

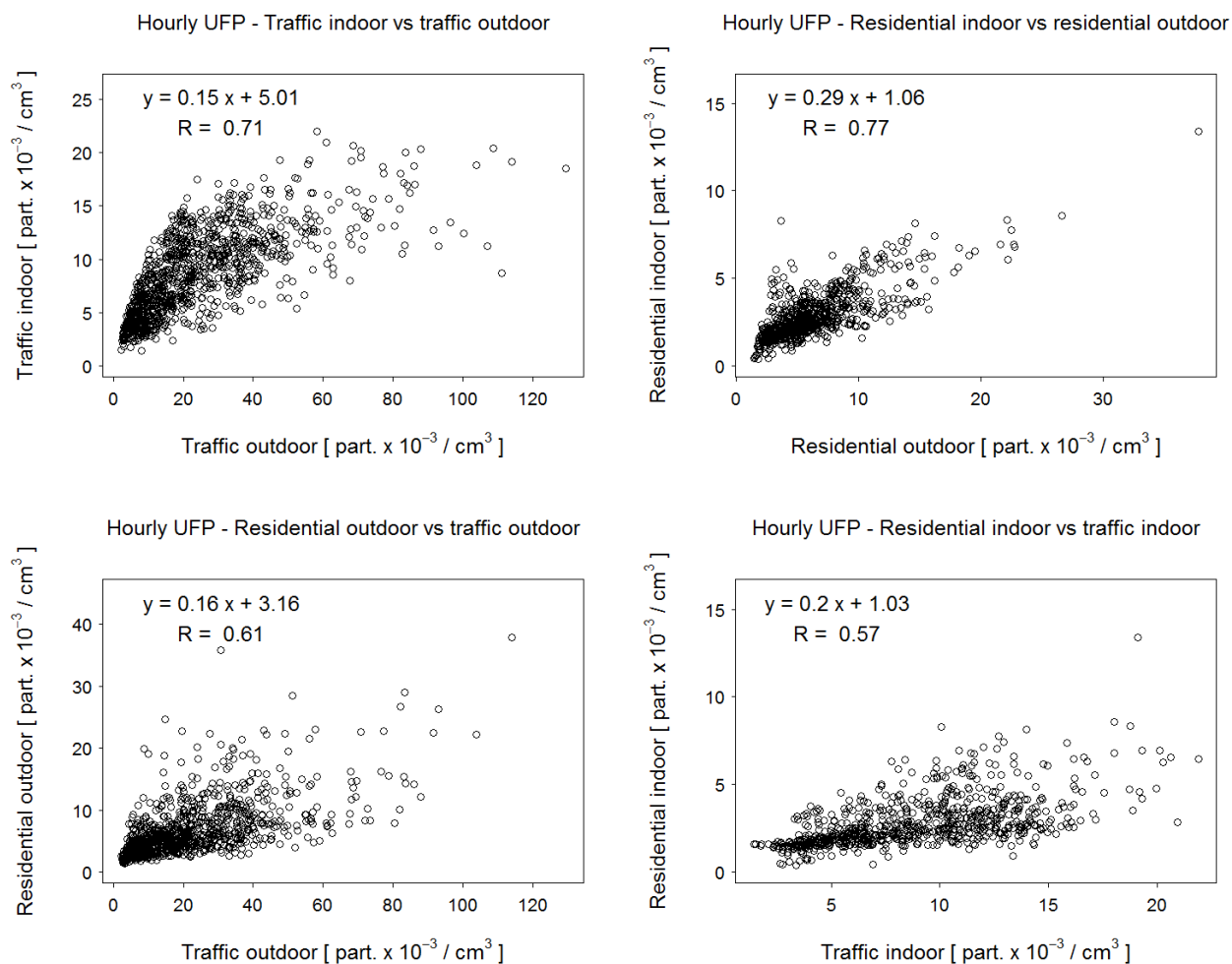


Figure 4. Scatter plot for hourly UFP number.

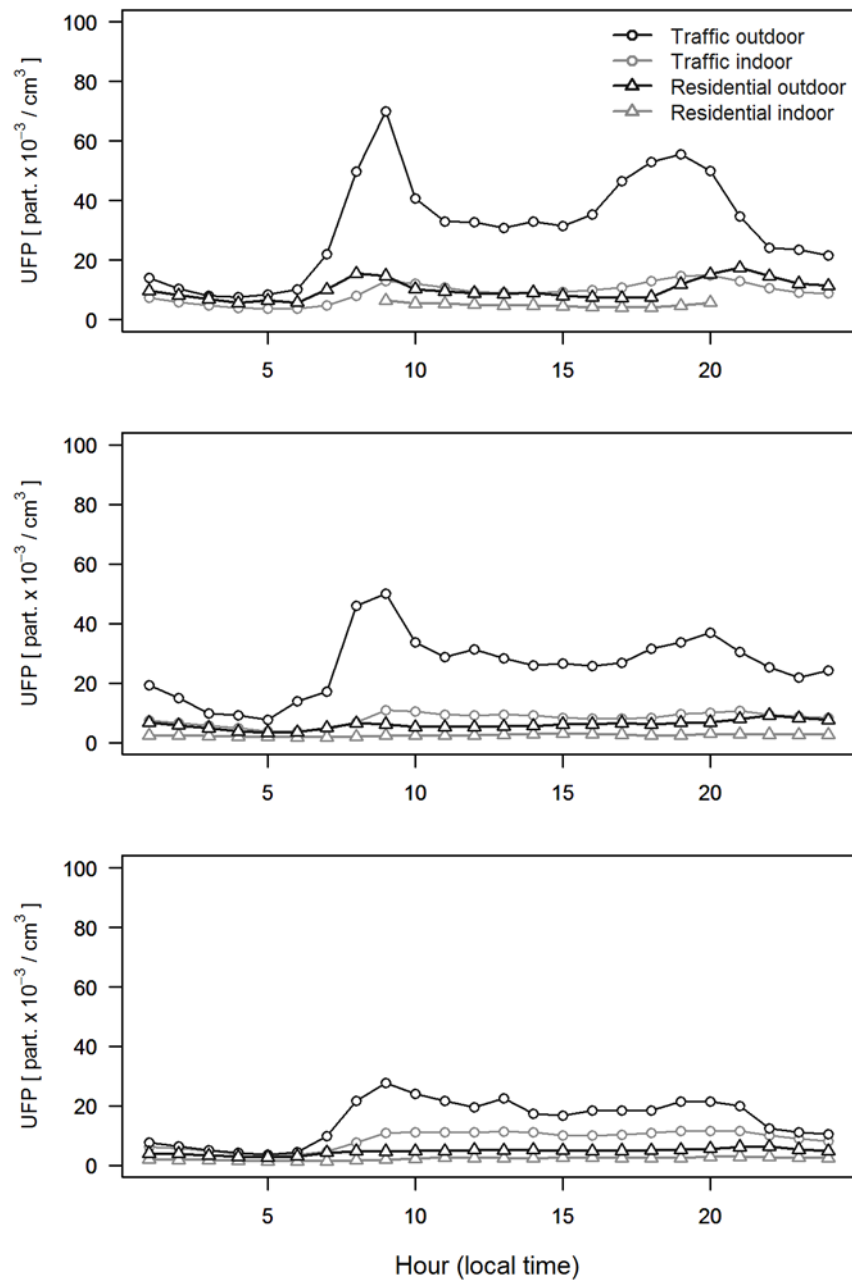


Figure 5. Daily temporal trend of UFP number during the three monitoring campaigns.

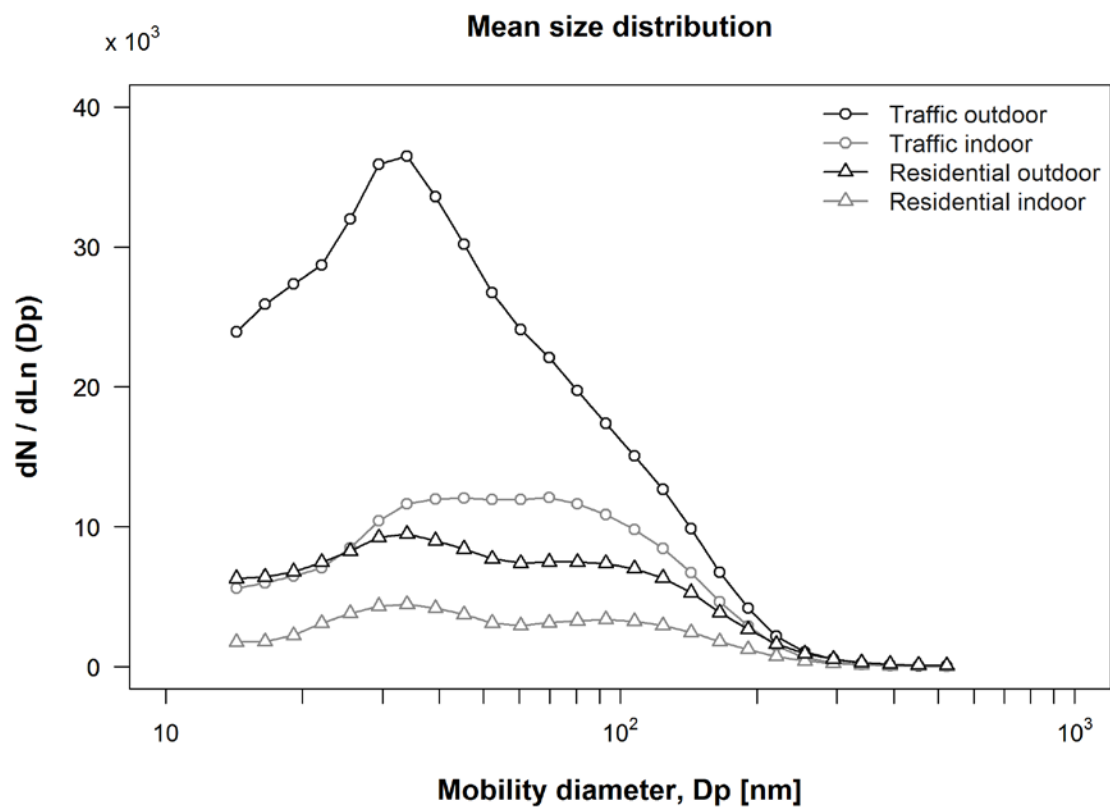


Figure 6. Mean particle size distribution of hourly data.

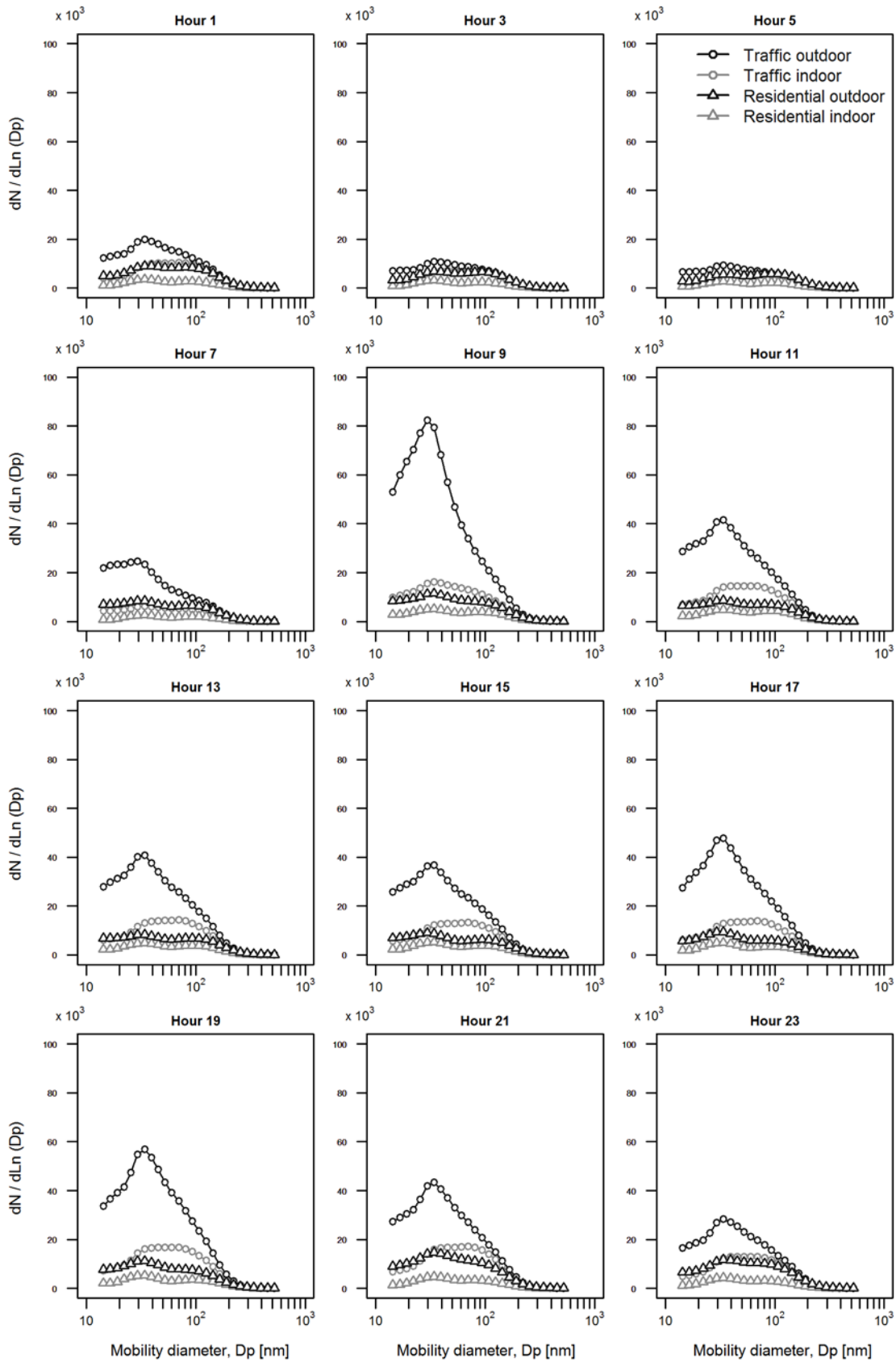


Figure 7. Mean particle size distribution of hourly data at specific time of the day.